## Deposition Temperature Mediated Selective Phase Transition Mechanism of VO<sub>2</sub> Films

Dooyong Lee<sup>1,2,\*,†</sup>, Donghyuk Yang<sup>1,\*</sup>, Hyegyeong Kim<sup>1</sup>, Jiwoong Kim<sup>1</sup>, Sehwan Song<sup>1</sup>, Kyoung Soon Choi<sup>2</sup>, Jong-Seong Bae<sup>3</sup>, Jouhahn Lee<sup>2</sup>, Jaekwang Lee<sup>1</sup>, Yunsang Lee<sup>4</sup>, Jiafeng Yan<sup>5</sup>, Jaeyong Kim<sup>5</sup>, and Sungkyun Park<sup>1,3,‡</sup>

<sup>1</sup>Department of Physics, Pusan National University, Busan 46241, Korea

<sup>2</sup>Advanced Nano Surface Research Group, Korea Basic Science Institute, Daejeon 34133, Korea

<sup>3</sup>Busan Center, Korea Basic Science Institute, Busan 46742, Korea

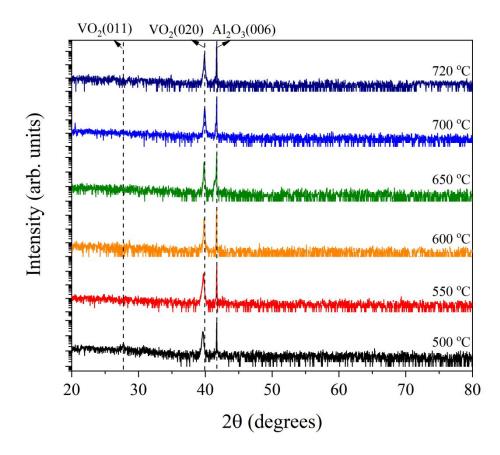
<sup>4</sup>Department of Physics, Soongsil University, Seoul 06978, Korea

<sup>5</sup>Department of Physics, Hanyang University, Seoul, 04763, Korea

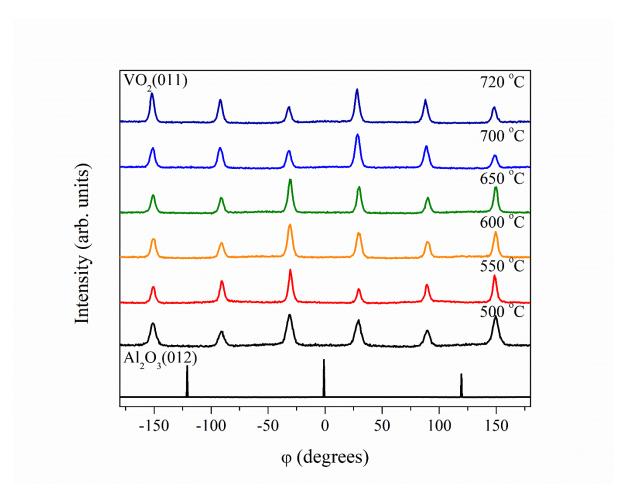
<sup>\*</sup> Both of them are equally contributed.

<sup>&</sup>lt;sup>†</sup> Current address: Department of Chemical Engineering & Materials Science, University of Minnesota,
Minneapolis, NM, 55455, USA

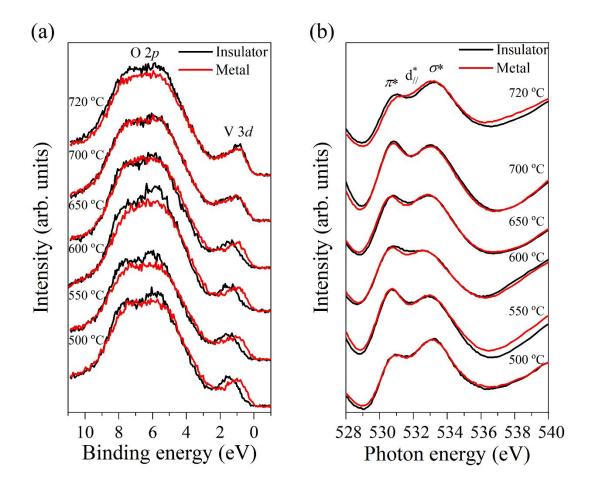
<sup>\*</sup> Correspondence should be addressed to Sungkyun Park (psk@pusan.ac.kr)



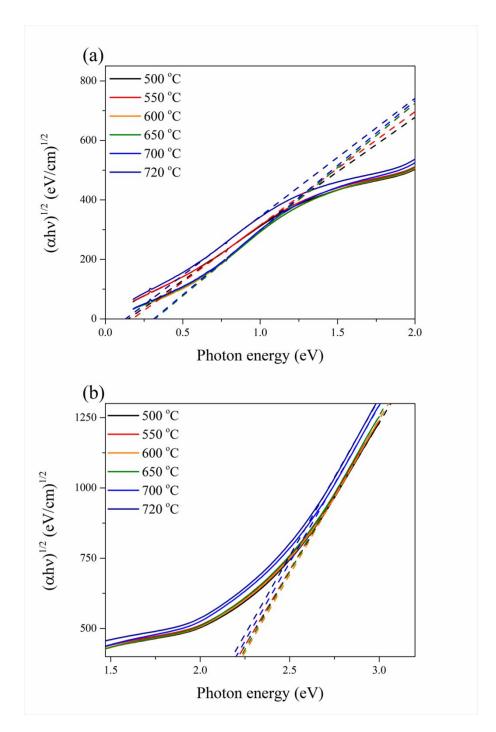
**Figure S1**. XRD  $\omega/2\theta$  full scans of VO<sub>2</sub> films grown on Al<sub>2</sub>O<sub>3</sub>(0001) substrates at various deposition temperatures. The vertical dotted lines indicate the reference Al<sub>2</sub>O<sub>3</sub>(006) (2 $\theta$  = 41.66°) and bulk monoclinic VO<sub>2</sub>(020) (2 $\theta$  = 39.88°).



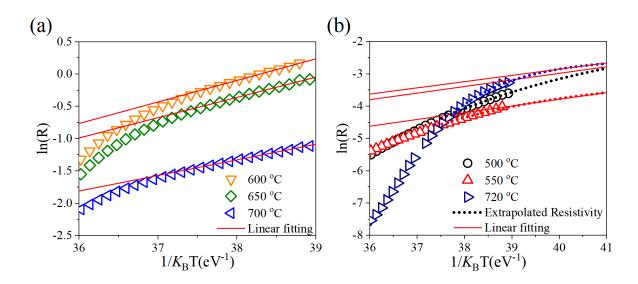
**Figure S2**.  $\varphi$ -scans of (011) plane of VO<sub>2</sub> films grown on Al<sub>2</sub>O<sub>3</sub>(0001) substrate at various deposition temperatures and the (012) planes of Al<sub>2</sub>O<sub>3</sub>(0001) substrates.



**Figure S3**. (a) Valence band spectra and (b) O K-edge XAS of VO<sub>2</sub> films deposited at 500, 550, 600, 650, 700, and 720 °C for the insulator (measured at RT) and metal (measured at 100 °C) phases.



**Figure S4**. Optical absorption spectra of  $VO_2$  films deposited at various temperatures in (a) low and (b) high photon energy region. Optical band-gap energy  $E_2$  ( $E_1$ ) is obtained from the absorption spectra in the low (high) photon energy region after linear extrapolation. Dashed lines represent the results of the linear fitting.



**Figure S5**. Plot of ln(R) vs.  $1/K_BT$  of  $VO_2$  films deposited at (a) 600, 650, and 700 °C and (b) 500, 550, and 720 °C. The solid curves indicate the liner fitted result to obtain the activation energy.

**Table S1**. Binding energy, FWHM and relative peak area ratio of deconvoluted V  $2p_{3/2}$  corelevel photoelectron spectra of VO<sub>2</sub> films with various deposition temperatures.

Deposition temperature (°C)	Binding energy (eV)			FWHM (eV)			Relative peak area ratio (%)		
	V <sup>3+</sup>	$V^{4+}$	$V^{5+}$	V <sup>3+</sup>	$V^{4+}$	$V^{5+}$	$V^{3+}$	$V^{4+}$	$V^{5+}$
500	515.20	516.14	517.43	1.50	1.31	1.39	2.54	59.25	38.42
550	515.20	516.16	517.44	1.50	1.37	1.47	2.46	59.25	37.97
600	515.20	516.17	517.47	1.50	1.37	1.45	2.64	61.78	35.57
650	515.19	516.18	517.49	1.50	1.37	1.39	3.98	62.54	34.38
700	515.20	516.17	517.43	1.50	1.23	1.40	6.78	58.22	35.00
720	515.19	516.19	517.47	1.50	1.28	1.42	8.62	57.03	34.35